Tracing the Steps in Nuclear Material Trafficking

Forensic scientists are combining an array of technologies to track illicit nuclear materials to their sources.

THE nuclear threat during the Cold War came from known adversaries, and a great concern for U.S. national security was that countries possessing a nuclear arsenal might use these weapons in a time of international crisis or by accident. The collapse of the Soviet Union and the increase in terrorism have given rise to new threats and adversaries. Today, the U.S. must prevent terrorists from acquiring not only nuclear weapons but also the materials that can be used to make such weapons, including fuel for nuclear power plants and radioactive materials intended for medical use.

In the last 10 years, a new research area called nuclear forensic analysis has emerged to address this growing concern. As part of the Laboratory's national security mission, researchers are assisting in an international effort to develop new methods to thwart illicit trafficking of nuclear materials.

Nuclear forensic science involves more than determining the physical and chemical characteristics of the nuclear material. Livermore physicist Ian Hutcheon, who serves as chief scientist for the Laboratory's nuclear forensic team, explains, "The goal is to identify as many clues in intercepted nuclear and radiological samples as possible. These so-called attribution indicators provide us not only with a characterization of the nuclear material, but also with environmental links that could help us trace the path the material took from its point of origin or determine how recently it passed from legitimate to illicit use."

Nuclear materials can be placed into one of three general categories: special nuclear material (SNM), reactor fuel, and commercial radioactive sources. SNM includes the basic building blocks of nuclear weapons: weapons-grade plutonium and highly enriched uranium (HEU). Reactor fuel includes low-enriched uranium (LEU), reactor- and fuel-grade plutonium, and mixed oxide– (MOX-) grade plutonium. (MOX fuel is a mixture of plutonium and depleted uranium oxides and can be a substitute for LEU.) Commercial radioactive sources are used in medical diagnostics, thermoelectric generators, food irradiators, and radiography equipment.

SNM is attractive to terrorists because, in sufficient quantities, it eliminates the need to enrich uranium or acquire plutonium. However, nations with nuclear weapon capabilities protect their HEU and plutonium stockpiles with extensive security systems. Reactor fuel cannot be directly used to build a nuclear device, but it is stored in many locations throughout the world and tends not to be as rigorously secured. Terrorists might try to acquire reactor fuel and other radioactive materials to construct a radiological dispersal device, or dirty bomb.

Livermore's Nuclear Forensic Roots

Nuclear forensic work at the Laboratory began in the late 1980s as an outgrowth of the nuclear test program. To analyze underground nuclear experiments, researchers mixed chemical and isotopic tracers with the nuclear test device and measured the isotopic composition of the debris produced during the experiment. These data would help scientists determine the device's yield and provide important feedback to the design team.

Livermore nuclear chemist Ken Moody believed trace radionuclides could be used in a similar fashion to determine the origin of a nuclear material. Radioactive atoms decay at a rate determined by the amount of the isotope in the material and the half-life of the parent isotope. For example, thorium-230 is a decay product, or daughter isotope, of uranium-234, and uranium-235 is a daughter of plutonium-239. Therefore, by measuring the relative amounts of decay products and the parent isotope, scientists can establish a material's age—the time since the parent isotope was last chemically separated from its decay products.

Moody explains, "If a sample is completely purified, every one of the daughters should have the same age. If they do not, the difference indicates that the material has been altered or been through chemical processing. Because each manufacturer has different processing methods, we can use the age and other isotopic information to help identify, or attribute, where a sample was manufactured."

Livermore physicist Sid Niemeyer submitted a proposal to the Department of Energy (DOE) for the Laboratory to conduct further research on these age-dating techniques. DOE then assigned Lawrence Livermore with responsibility for leading the national laboratory effort in nuclear forensic analyses. In 2003, recognizing Livermore's technical expertise in nuclear forensic science, the Department of Homeland Security (DHS) selected the Laboratory to lead a new national program in nuclear attribution of undetonated materials.

Incidents of illegal trafficking in nuclear materials are not new. A DOE database lists worldwide cases from as early as 1966. However, until recently, these cases were usually frauds—that is, attempts by individuals to sell substances they claimed were nuclear material. Since 1991, the number of reported nuclear smuggling cases has risen, with approximately one seizure per month. Most of the seizures have involved small amounts of nuclear material produced in commercial nuclear reactors.

Many of the techniques used in traditional criminalistics are also useful for nuclear forensic analysis, such as microscopic analyses of fibers and packaging materials. However, in nuclear forensic analysis, radioactive elements are being analyzed, so the procedure used depends on the type of sample and its radioactivity. Forensic scientists must determine, for example, whether a sample is emitting large amounts of alpha particles or gamma rays; whether it is plutonium or uranium; or whether it is a solid, mixed powder, or liquid. Therefore, Livermore's nuclear forensic team uses a tiered approach in which the results from one analysis guide the team in selecting subsequent analyses.

Identifying the Nuclear Material

Every radioactive sample that arrives at the Laboratory's Forensic Science Center (FSC) is first screened for alpha particles and gamma rays. (See *S&TR*, May 2003, pp. 4–11; April 2002, pp. 11–18.) If a substantial amount of either is emitted, the sample is analyzed using sensitive spectrometers designed to measure those particles. Once researchers identify the material, they analyze its chemical and isotopic composition, which includes measuring the amount of major and trace elements in the sample, evaluating its physical structure, and in particular, determining the ratio of radioactive parent isotopes to daughter isotopes.

Livermore forensic scientists pioneered the use of this tiered approach, in which nondestructive techniques such as optical and scanning electron microscopy, Fourier-transform infrared (FTIR) spectrometry, and x-ray fluorescence (XRF) are first applied to determine a sample's overall composition and structure. The complete characterization of a sample by radiochemical and mass spectrometric analysis involves dissolution or vaporization, which destroys the sample. Therefore, these techniques are the final steps in the forensic process.

Livermore researchers work with other DOE national laboratories in forensic investigations, including Los Alamos, Oak Ridge, Pacific Northwest, and Savannah River national laboratories. Typically, Livermore performs a preliminary analysis of a nuclear sample. Then a second confirming analysis is made to ensure accuracy. Oak Ridge conducts this second analysis if the sample is uranium, and Los Alamos performs the analysis if the sample is plutonium. Oak Ridge also lends its expertise in x-ray diffraction, and Pacific Northwest and Savannah River provide expertise in nuclear reactor design and fuel reprocessing.

Because the nuclear forensic field is new and has relatively few experts, countries engaged in nuclear attribution research often turn to the international forensic science community for technical

The Nuclear Smuggling International Technical Working Group, formed in 1995, works closely with the International Atomic Energy Agency to provide assistance to nuclear forensic scientists around the world. In 2004, the group, which included participants from Lawrence Livermore, met in Cadarache, France, to discuss recent progress in countering the illicit trafficking of nuclear materials.

assistance. In 1995, Niemeyer and Lothar Koch, former division leader at the Institute for Transuranium Elements in Karlsruhe, Germany, started the Nuclear Smuggling International Technical Working Group (ITWG). Experts from 28 countries meet once each year to work on issues concerning illicit trafficking of nuclear materials. The group's objectives include developing protocols for collecting evidence, prioritizing techniques for forensic analyses of nuclear and associated nonnuclear samples, conducting interlaboratory forensic exercises, and developing forensic databanks to assist in interpretation.

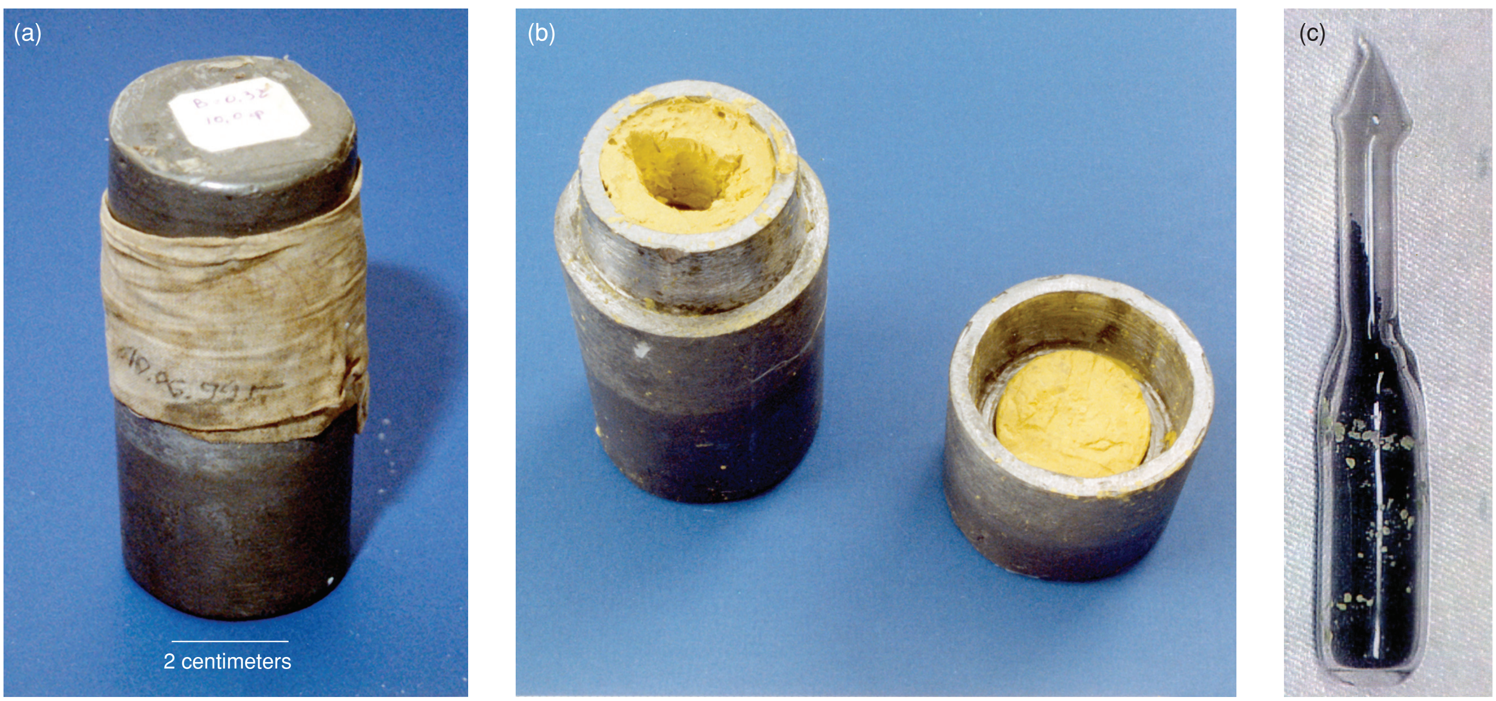
"We want the working group to be a clearinghouse for scientific information in the nuclear forensic field," says Livermore geochemist Dave Smith. ITWG works closely with the International Atomic Energy Agency (IAEA) to provide requesting countries with forensic analyses and support. In 2004, ITWG organized the International Nuclear Forensic Laboratories to establish guidelines for best practices, conduct international exercises, promote research and development, publish reports, and provide point-of-contact assistance.

Countries possessing the equipment necessary to conduct forensic analyses, such as the United Kingdom, France, or Germany, perform their own analyses if nuclear material is intercepted within their borders. Countries without the necessary equipment, for example, many of the smaller nations in Eastern Europe, usually turn to the U.S. for nuclear attribution assistance.

For example, in May 1999, Bulgarian customs officers detained a man who had hidden a 2.4-kilogram lead container in the trunk of his car. Documents found on the suspect described the material as 99.99 percent uranium-235. Inside the container was a glass ampoule filled with several grams of fine black powder. The ampoule was wrapped in paper and cushioned by yellow wax. (See the figure below.) The U.S. Department of State arranged for the National Academy of Sciences in Sofia, Bulgaria, to send the container to Livermore for analyses. The results indicated that the material originated from Eastern Europe.

Nuclear Tracing, Step by Step

Part of the protocol for handling samples sent to the FSC is to set up and maintain a chain-of-custody record. The FSC provides an important link between Livermore's nuclear scientists and federal and state law enforcement agencies. Established in 1991, the center supports DOE in verifying compliance with international treaties, and in 1998, FSC researchers developed methods to help verify the safety of weapons in the U.S.



(a) A lead container shielding an ampoule filled with uranium was seized at the Bulgarian border in 1999 and sent to Livermore for analysis. (b) The interior of the container was lined with yellow wax, which was used to cushion the nuclear material. (c) The glass ampoule contained almost 4 grams of highly enriched uranium oxide.

nuclear stockpile. The team has also assisted federal, state, and local law enforcement on a wide range of cases, including the 1993 World Trade Center bombing and Unabomber investigations.

"The best way to test the techniques we use is to work on real cases," says Livermore chemist Pat Grant, who is the FSC deputy director. "They help us improve our methods and sometimes lead us to develop new techniques."

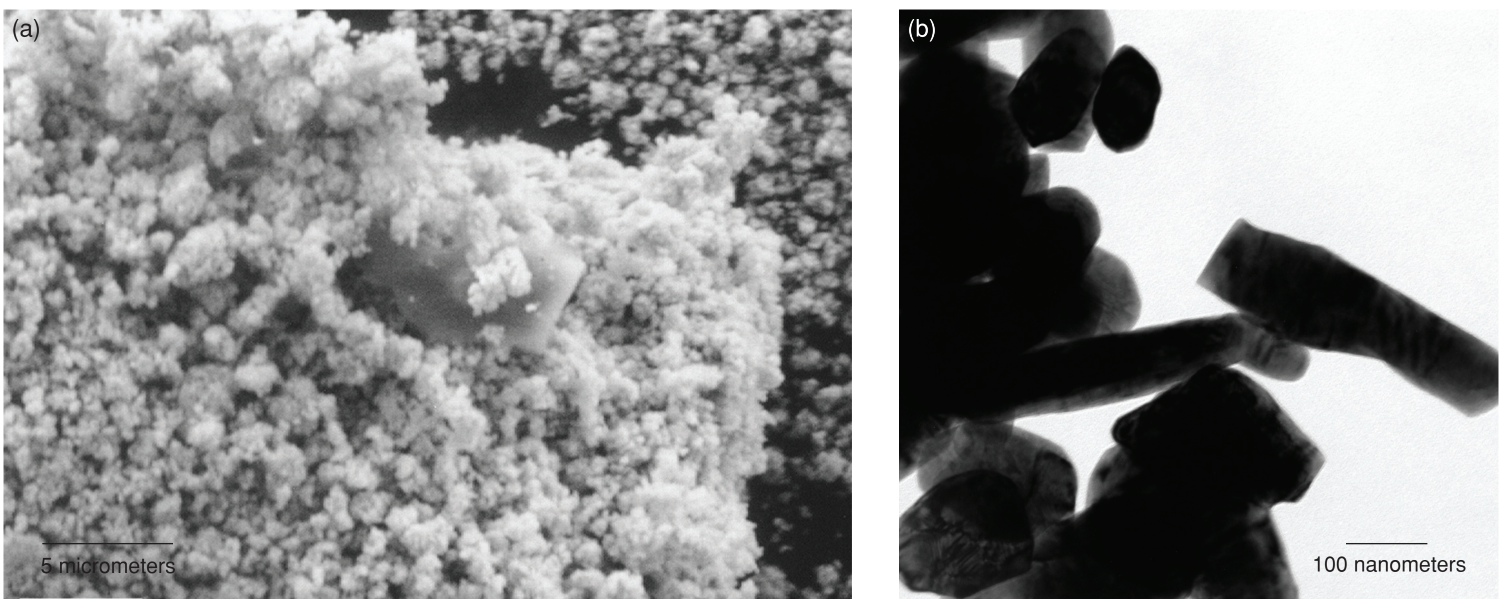
Livermore has had a memorandum of understanding in place with the Federal Bureau of Investigation since 1998 to assist the bureau in combating terrorism. One FSC project develops methods to ensure the integrity of conventional forensic evidence should a dirty bomb be detonated. Residue from such an explosion would contain signatures that might provide important clues to a material's origin. Signatures of a given nuclear or radiologic material include the physical, chemical, and isotopic characteristics that distinguish it from other nuclear or radiological materials. They enable researchers to identify the processes used to create a material. After a detonation, however, hair, fibers, and other evidentiary material near the blast would become contaminated with radioactive material. The challenge for scientists is to preserve the conventional evidence while separating out any radioactive contaminants.

FSC scientists have learned that analyzing the materials accompanying a radioactive sample is as important as characterizing the sample. These so-called route materials—such as containers, fingerprints, fibers, and pollen—provide attribution details about who has handled a sample or the path it has traveled. In the Bulgarian seizure, for example, Livermore scientists used FTIR to confirm that the yellow wax was paraffin. XRF results indicated the yellow coloring was barium chromate, an additive rarely used in Western countries because of environmental concerns but commonly used in Brazil, China, India, and Eastern Europe. Optical microscopy of the paper surrounding the ampoule and the label on the container showed that both of these were a mixture of hardwood and softwood tree fibers commonly found in Eastern Europe.

Tools of the Trade

The FSC works closely with Livermore's Chemistry and Materials Science (CMS) Directorate, which supports a vast array of instruments that researchers can use to build a physical, chemical, and isotopic profile of a material sample. Two tools commonly used in nuclear forensic work are the scanning electron microscope and the transmission electron microscope, which can provide detailed images at resolutions better than 6 and 0.5 nanometers, respectively. Such high-resolution images allow researchers to study the microstructure and chemical composition of individual nanometer-size particles. (See the figure below.)

Because a powdered sample can contain material with different shapes, sizes, and textures, imaging tools must completely characterize a sample while ensuring that important signatures can be recovered from individual components. This step is an essential prerequisite to chemical and isotopic characterization, where components are separated for individual analysis. Moody, who performs radiochemical



(a) This high-resolution image was taken with a scanning electron microscope. The smooth-surfaced, angular object in the center is a shard of glass from an ampoule that contained uranium oxide. (b) The sample was also analyzed with transmission electron microscopy, allowing researchers to study the material's microstructure.

analyses on nuclear samples, relies on such characterization results to help determine which chemicals are needed to break each sample down into a solution for age dating. Chemical analysis reveals the material's exact chemical composition or the association of unique molecular components.

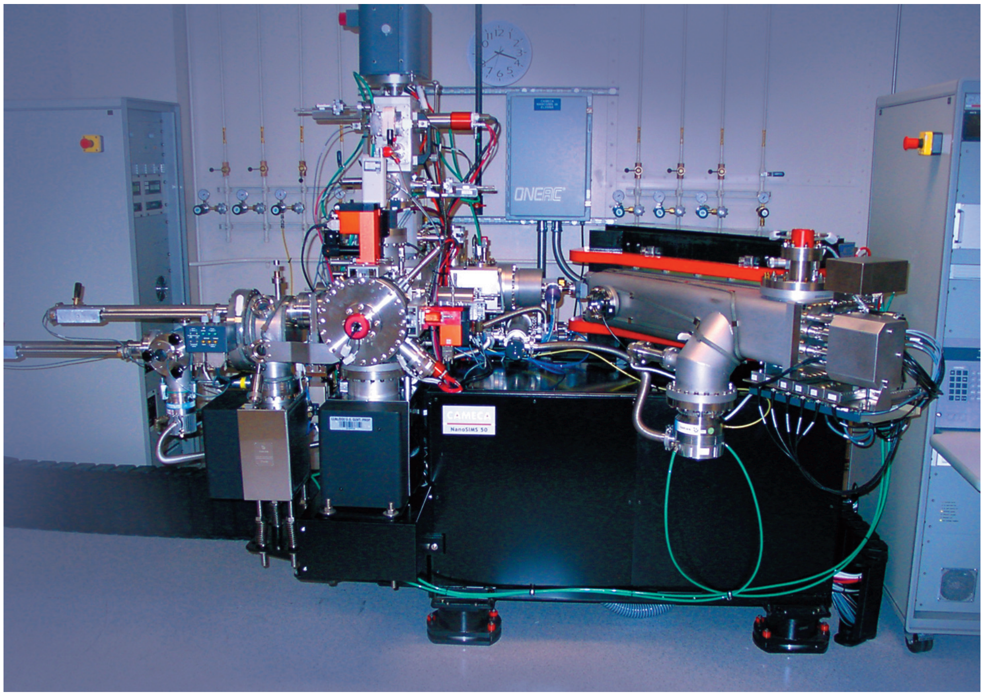
For example, electron microscopy and x-ray diffraction showed the Bulgarian sample to be uranium oxide. Uranium oxide comes in many forms, each of which can be found at various points in the uranium fuel cycle. Trace elements and organic compounds associated with the nuclear material are also important indicators of a material's history.

Another tool widely used in nuclear forensic analysis is the mass spectrometer. In mass spectrometry, a sample is converted into positively and negatively charged ions. The ions are then separated according to their mass-to-charge ratios, and the intensities of the separated ion beams are measured. The secondary ion mass spectrometer (SIMS) can be used as both a microscope and a microprobe. At present, it is the only technique that can perform sensitive elemental and isotopic analysis with submicrometer spatial resolution. Results can be obtained from a specimen as small as a few picograms (10^{-12} grams), about one-millionth the mass of a grain of sand.

The Laboratory's NanoSIMS (shown in the bottom right photograph) is capable of nanometer-scale resolution. Lawrence Livermore is the only national laboratory with this spectrometer, which can also be applied to biological materials analysis, microbial mineralization processes, molecular targeting for cancer therapy, and cosmochemistry research.

The FSC also uses mass spectrometry to analyze the chemistry of route materials. Gas chromatography–mass spectrometry (GC/MS) is useful for measuring trace organic constituents in bulk samples and can analyze samples as small as a grain of salt. In GC/MS, a sample is injected into the end of a column through which gas is flowing. When the sample is heated, it

Livermore nuclear chemist Ken Moody performs forensic radiochemical analysis on samples inside a glovebox. The procedures for working with radioactive samples ensure a safe working environment and maintain the integrity of the samples.



Livermore's newest secondary ion mass spectrometer, called NanoSIMS, can perform ultrasensitive elemental and isotopic analyses at nanometer-scale resolution.

vaporizes into an aerosol of the sample's chemical components. Because chemicals have different vapor pressures and chemical affinities, each component migrates down the column at a different rate. After the components exit the column, they are bombarded with an electron beam, which causes the molecules to break apart into fragment ions and produces a material fingerprint that identifies the compound.

Cataloging the Results

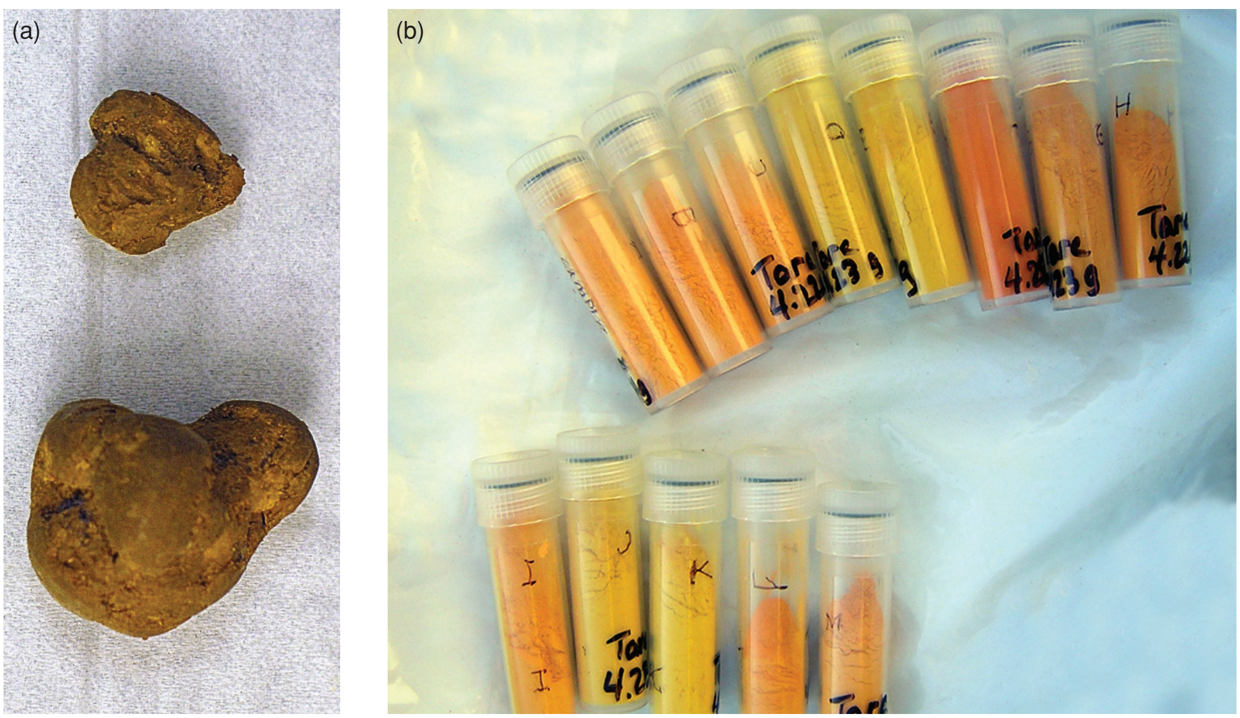
The complete analyses of the Bulgarian sample revealed that it was 73 percent uranium-235 and 12 percent uranium-236, consistent with material that had been recycled from very highly enriched nuclear reactor fuel. Although the amount of HEU confiscated was far short of the quantity needed to fashion a crude bomb, the level of enrichment placed the material in the weapons-grade category.

This incident underscores the security concerns involved in operating commercial and research nuclear reactors. Commercial reactors store large quantities of irradiated nuclear fuel in cooling ponds exposed to the open air. Research reactors often lack containment structures and exclusion zones, and they may be located on university campuses or in other densely populated areas. According to a recent IAEA report, about 130 research reactors in 40 countries operate on weapons-usable HEU.

Nations with nuclear capabilities are beginning to share information about their nuclear processes and materials. Validating the signatures, measurements, and interpretive tools is an important part of building on the Laboratory's nuclear attribution capabilities. Progress in this area requires what the Livermore team calls knowledge management. To compare a sample's signature with known signatures from nuclear production, reprocessing, manufacturing, and storage, researchers must first assemble a library of nuclear materials of known origin. The Livermore team is building its library with the help of Westinghouse and other domestic nuclear fuel fabricators by obtaining samples and data to serve as standards.

Knowledge management also includes determining the best ways to archive and retrieve information, especially during emergencies. According to Smith, knowledge management is as important as analysis capabilities, and it is an area of nuclear attribution that needs much work. To help Livermore improve its capabilities in this area, a nuclear engineer recently joined the team to manage a new attribution database of nuclear signatures.

The database approach will be useful in addressing all WMD issues. Hutcheon explains, "In cases involving potential WMD threats, we are addressing the same questions: How was the sample developed? Where did it originate? How old is it, and what is its intended use? We also want to be able to identify and correct any failures in our tracking system."



(a) A sample of uranium oxide is shown before its chemical composition has been analyzed. (b) A suite of known uranium oxide samples are used in comparison studies.

The Laboratory is involved in knowledge management activities across the spectrum of nuclear, chemical, and biological threats. For example, in 2004, Livermore established the Biodefense Knowledge Center to provide DHS with on-call technical assistance in the fight against bioterrorism. The center draws on about 75 researchers based at four national laboratories—Oak Ridge, Pacific Northwest, Sandia, and Lawrence Livermore.

According to Smith, ITWG also plans to develop databases as an international resource for nuclear forensic work. Elements of such databases already exist, but most stored data are collected for environmental impact analyses, weapons performance testing, or other purposes. Although the information may be valuable for nuclear forensic analysis, no country is likely to divulge all of the information in its databases. Thus, ITWG's goal is to serve as an intermediary, where countries can confirm results or fill in missing pieces of information, if presented with details of a suspicious material.

Programs to Safeguard Material

Livermore researchers are also contributing to projects to help Russia secure, consolidate, and eliminate fissile materials. (See *S&TR,* January/February 2005, pp. 14–21.) In May 2004, then Secretary of Energy Spencer Abraham launched the Global Threat Reduction Initiative (GTRI), with the goal of repatriating all Soviet-origin HEU fuel to Russia by the end of 2005. DOE plans to work with Russia to repatriate all Soviet-origin spent nuclear fuel by 2010 and all spent research reactor fuel of U.S. origin by 2015. The U.S. government has announced plans to dedicate more than $450 million to the GTRI, and an organization has been created within the National Nuclear Security Administration to implement this and other GTRI tasks. A complementary effort has been the construction of the Mayak Fissile Material Storage Facility at Ozersk, Russia, a program sponsored by the Department of Defense. When completed, the facility will house and secure 50 tons of Russian weapons-grade plutonium.

Other Laboratory research supports DHS in its efforts to screen for radioactive materials at the nation's border security checkpoints. One area of concern is that radioactive materials could be hidden on ships destined for U.S. ports. To address this problem, DHS has funded the Container Security Initiative, which focuses on securing the ports that handle nearly 90 percent of the world's shipping. One Livermore project in support of this initiative combines the Laboratory's expertise in radiation science and detection to quickly screen cargo containers. (See *S&TR*, May 2004, pp. 12–15.)

Tomorrow's Nuclear Scientists

At a time when the nation has a critical need for nuclear forensic scientists, the number of students at U.S. universities who are pursuing nuclear science degrees is declining. A primary reason for this reduced focus is the declining nuclear power industry and the lack of job opportunities for graduates. The country's research reactors are being closed, and new commercial nuclear reactors are not being built. Livermore's nuclear forensic team wants to reverse this educational trend and is working with the Laboratory's Glenn T. Seaborg Institute, which is part of CMS, to identify and attract the next generation of nuclear scientists.

Annie Kersting, the institute's director, says, "We see nuclear attribution as a new career avenue for students. Working with our equipment is an attractive option for students interested in nuclear science and will help the Laboratory to rebuild the nuclear chemistry expertise we had in the days of underground testing."

Tomorrow's generation of weapons scientists face many new and unexpected challenges in addressing the world's nuclear threat. As Livermore continues to ensure the performance and safety of the U.S. nuclear stockpile—a vital part of its national security mission—nuclear forensic scientists will continue to develop tools and attribution processes to trace illicit nuclear materials to their sources.

—*Gabriele Rennie*

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